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We have measured the specific heat, resistivity, and ac susceptibility of (K, Ba)BiO_3 single crystals before and after introduction of either point or columnar defects by electron (EI) or heavy-ion irradiation (HII). While the magnetic field dependence of these properties remains mainly unaffected by EI, the irreversibility line and the location of the specific heat anomaly are both shifted up in temperature after HII. The shift is apparent only if the magnetic field is applied parallel to the ion tracks. For perpendicularly applied fields, both lines lie at the same field as in the pristine sample. These experiments call the nature of the vortex liquid state into question.

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The vortex phase diagram of high T_c superconductors (HTS) has proven to be a remarkably complex and rich area of study. One of the most extraordinary discoveries has been the melting of the flux line lattice to a vortex liquid phase [1]. The large extent of the latter phase in the (H, T) phase diagram is believed to be the result of thermal fluctuations, which in HTS are enhanced by the combination of high critical temperature and large crystalline anisotropy. However, transport measurements and neutron diffraction on the vortex lattice in the fully *isotropic* (i.e., cubic) (K, Ba)BiO_3 (KBBO) system with $T_c \sim$ 30 K yield results that are very similar to the phenomenology of HTS [2,3]. Those measurements are suggesting the existence of a vortex liquid over a large portion of the phase diagram in this material, too.

On the other hand, recent tunneling and specific heat measurements [4] (C_p) have shown that the *superconducting* transition, deduced from the specific heat jump, occurs significantly *below* the temperature at which the superconducting gap opens in the tunneling data, and that this transition is closely related to the irreversibility line (IRL, at which $\rho \rightarrow 0$). Indeed, both thermodynamic (deduced from C_p) and dynamic (IRL) lines show an unexpected positive curvature calling into question the nature of the transitions in KBBO. The question of the existence of a transition in the vortex state well below the upper critical field $H_{c2}(T)$ is not only particularly pressing in cubic (Ba, K)BiO_3 but also in single-layer cuprates such as (Nd, Ce)CuO_4 [5] and $Tl_2Ba_2CuO_6$ [6]. Unlike other HTS materials, single crystals of these materials do not show any broadening of the resistive transition above the irreversibility line, which first lead to the latter's identification with H_{c2} [6]. This issue remains open as later specific heat measurements in $Tl_2Ba_2CuO_6$ suggested that the H_{c2} line lies well above the IRL [7].

In order to clarify the role of quenched disorder as opposed to thermal fluctuations on the superconducting transition, we have investigated the effects of both electron (EI) and heavy-ion irradiation (HII) on the resistivity, specific heat, and ac screening of single crystalline (K, Ba)BiO_3. The central result is that, after the introduction of columnar defects by HII, the IRL *and* the thermodynamic superconducting transition line $T_{C_p}(H)$ deduced from specific heat measurements *both* shift towards higher temperature when the external field H is applied parallel to the defects. Both lines move back to the position observed in the pristine sample when the magnetic field is applied perpendicular to the track direction. Neither line is affected by EI. An upward shift after HII has been predicted only for the transition from the vortex liquid to the Bose glass (BG) [8]; its observation by calorimetric measurements calls the existence of a distinct vortex liquid phase into question.

The samples studied here were very homogeneous single crystals, from the same batch, and close to optimal doping ($T_c \approx 31.5$ K). Sample 1 was kept pristine, sample 2 was irradiated with 4.6×10^{19} cm^{-2}, 2.5 MeV electrons, and sample 3 was irradiated with 7.2 GeV Ta^{57+} ions at the Grand Accélérateur National d'Ions Lourds (Caen, France). The ion irradiation dose, $n = 3 \times 10^{11}$ ions cm^{-2}, corresponds to a dose-equivalent matching field $B_\Phi \equiv \Phi_0 n = 6$ T ($\Phi_0 = h/2e$ is the flux quantum). Transmission electron micrographs have shown that HII results in the formation of linear amorphous tracks with diameter $c_0 \sim$ 6–8 nm [9]. The vortex matter in heavy-ion irradiated (K, Ba)BiO_3 closely obeys the "Bose-glass" phenomenology [9], in which the vortices become localized due to their interaction with the columnar defects [8]. This behavior is again, with surprising detail, similar to that found in much more anisotropic $YBa_2Cu_3O_7$ after heavy-ion irradiation [10].

The specific heat was measured by an ac technique [11] that allowed us to measure small samples (here, a few 10^{-2} mm^3) with high sensitivity (typically one part in 10^4). For the transport measurements, electrical contacts were prepared by applying strips of silver paint on the sample surface after irradiation and subsequently annealing the sample at 500 °C for a few hours. We have measured the I-V characteristics (where I is the current and V the voltage) and the resistivity as a function of temperature with a resolution of 0.5 nV. The third harmonic ac response, which detects the nonlinearity of the $I(V)$ curve at very low voltage, was studied by measuring the local magnetic transmittivity using a miniature Hall probe [12].

Figure 1 displays the temperature dependence of the specific heat at various magnetic fields up to 5 T in the pristine (top panel) and heavy-ion irradiated samples (bottom panel). The curve measured at $H = 7$ T coincides with the normal state specific heat above $T_c(7\ \mathrm{T})$ and has been used as a baseline. $T_c(7\ \mathrm{T}) \approx 20$ K for the pristine sample and ≈ 24 K in the irradiated sample (for H parallel to the tracks), and only the data above $T_c(7\ \mathrm{T})$ are presented in Fig. 1. Since our measurements are only relative, the data were normalized using those of Ref. [13]. In spite of the elevated track density, which corresponds to an amorphization of 15% of the material, the irradiated crystal shows no significant shift of the critical temperature ($T_c \sim 31.3$ K) nor any increase of the superconducting transition width, which excludes possible effects of pair-breaking or self-doping [14]. The specific heat anomaly remains well defined over the whole investigated magnetic field range and the amplitude of the jump drops by about 15% after HII as expected from the presence of 15% of amorphous nonsuperconducting phase in the columnar tracks. However, as shown, the shift of the specific heat anomaly with magnetic field is *much smaller* after heavy-ion irradiation.

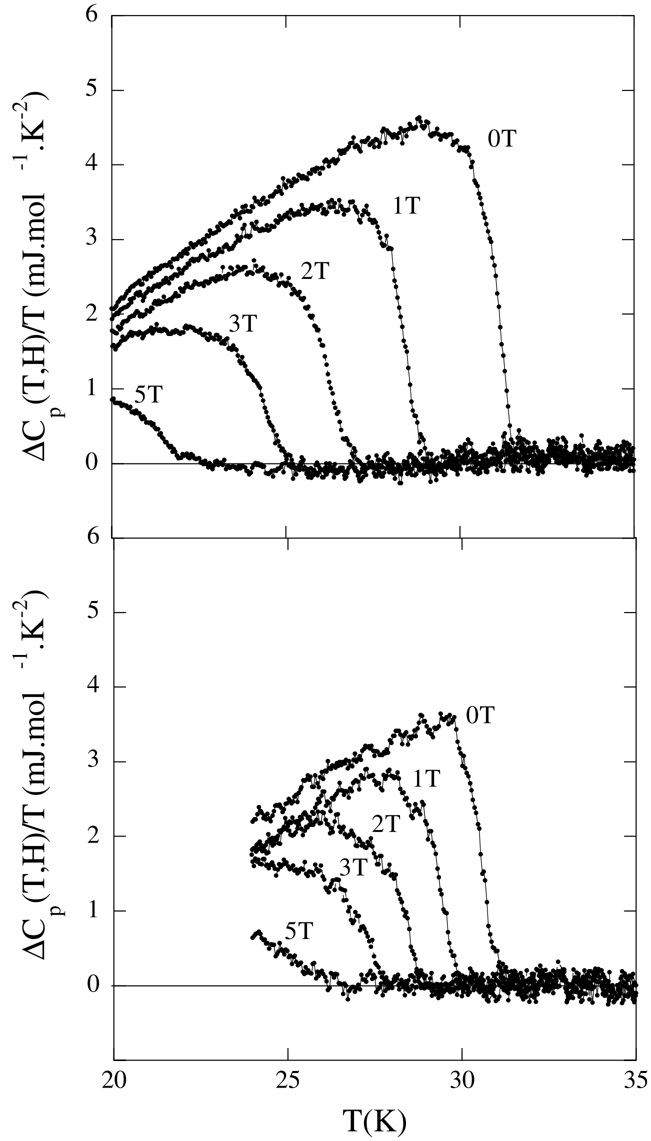

FIG. 1. Temperature dependence of the specific heat anomaly in a pristine $(K, Ba)BiO_3$ single crystal (top panel) and after irradiation with 7.2 GeV Ta ions for $H||$ to the columnar tracks ($B_\Phi = 6$ T, bottom panel).

Figure 2 shows the $H_{C_p}(T)$ curve defined as the onset of the specific heat anomaly, together with the IRL, $H_{\mathrm{irr}}(T)$, deduced from the ac transmittivity measurements (and which corresponds to the temperature for which $\rho \to 0$ in transport measurements). Note that $T_{\mathrm{irr}}(H)$ approximately corresponds to the midpoint of the specific heat anomaly. EI, which produces Frenkel pairs (point defects), results in a notable decrease of the critical temperature to $T_c \approx 29.2$ K, but without any significant broadening of the transition. Despite the reduction of T_c, the magnetic field dependence of the specific heat anomaly is very similar to that of the pristine sample, if plotted as a function of T/T_c. Only a slight downward shift of the IRL is observed, consistent with the introduction of point defects which are expected to promote vortex line wandering [15].

On the contrary, for a magnetic field applied parallel to the columnar defects, the heavy ion-irradiated crystal remarkably displays a marked, similar upward shift of both H_{C_p} and H_{irr} lines. Moreover, if the field is applied perpendicular to the ion tracks, the specific heat anomaly moves back to the position measured in the pristine crystal (see Fig. 3) [16]. Even more remarkably, this shift is completely consistent with the one observed in the resistive transition. Both curves lie at a higher temperature than in the pristine crystal when H is parallel to the ion tracks, but shift back to their original position when H is applied perpendicular to the tracks. However,

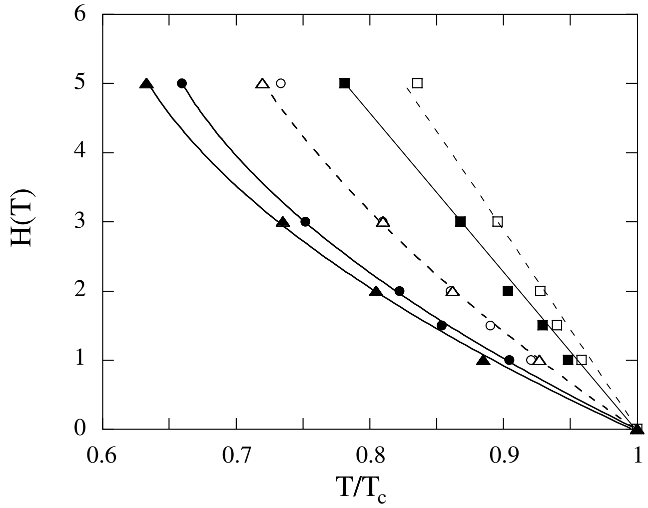

FIG. 2. (H, T) phase diagram deduced from specific heat (open symbols) and third harmonic ac screening measurements (solid symbols) on a pristine (circles), a heavy ion irradiated ($B_\Phi = 6$ T, squares), and an electron irradiated (4.6×10^{19} cm^{-2}, 2.5 MeV electrons, triangles) (K, Ba)BiO$_3$ single crystal. For the heavy-ion irradiated crystal, the magnetic field was applied parallel to the ion tracks.

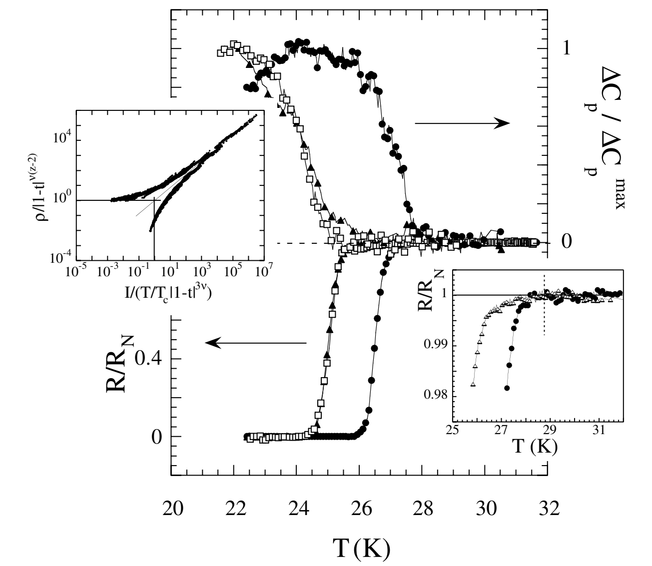

FIG. 3. Specific heat anomaly and resistivity vs temperature in a pristine crystal (squares) and a HII (K, Ba)BiO$_3$ ($B_\Phi = 6$ T), with field ($\mu_0 H = 3$ T) applied both parallel to (solid circles) and perpendicular to (triangles) the tracks. Upper inset: Bose-glass scaling of the I-V characteristics at different temperatures in magnetic fields ($\parallel$ to the tracks, $t = T/T_{\rm irr}$) $\mu_0 H = 3\ {\rm T} = \frac{1}{2}B_\Phi$ and $\mu_0 H = 7\ {\rm T} \sim B_\Phi$. Lower inset: expanded view of the top of the resistive transition showing that its onset temperature is independent of the direction between the tracks and the field; $H \parallel$ tracks: circles; $H \perp$ tracks: triangles.

the temperature $T_{\rm on}$ at which fluctuation paraconductivity becomes measurable (i.e., the lowest temperature at which R coincides with the normal state resistivity R_N) is *independent* of field orientation, and equal to $T_{\rm on}$ measured in the pristine sample (lower inset of Fig. 3). In the latter case, $T_{\rm on}$ corresponds closely to the temperature T_Δ at which the superconducting gap completely disappears [17].

In the presence of columnar defects, the foot of the resistive transition is usually described [18] by scaling functions describing the establishment of long-range phase coherence at the vortex liquid to Bose-glass transition temperature $T_{\rm BG}$ [8]. This transition can then be characterized by two critical exponents ν and z which describe the divergence of the phase correlation lengths $\xi_\parallel$ parallel to the defects and the time scale τ on which those fluctuations can survive [8] (for screened vortex interactions, the correlation volume is anisotropic and $\xi_\perp$ perpendicular to the columns obeys $\xi_\parallel \sim \xi_\perp^2$). The upper inset of Fig. 3 shows typical scaling of the $I(V)$ characteristics in our samples (at $B = 3\ {\rm T} = \frac{1}{2}B_\Phi$ and $B = 7\ {\rm T} \gtrsim B_\Phi$). This formalism leads to $z \sim 4.8 \pm 0.4$ and $\nu \sim 1.1 \pm 0.2$ in good agreement with numerical simulations for the BG transition [19].

The introduction of columnar ion tracks impedes vortex motion and line wandering, thereby promoting phase coherence in the mixed state [20]. The upward shift of the IRL after HII (and its slight decrease after electron irradiation), its shift back to the position observed in the pristine sample when the magnetic field is applied perpendicular to the direction of the tracks, and its progressive increase with increasing ion fluence [9] seem to justify its identification as a transition from a vortex solid to a vortex liquid [21]. Such a transition is further confirmed by the scaling of the I-V characteristics. The puzzling result of this Letter is that the *thermodynamic* superconducting transition deduced from specific heat measurements is shifted consistently with the IRL. As observed for the IRL, preliminary measurements on a crystal with $B_\Phi = 1$ T show that the $H_{C_p}(T)$ line also progressively shifts towards higher temperature as the irradiation dose is increased.

The resistivity of our samples is approximatively $\approx 100\ \mu\Omega$ cm at T_c, and the Hall coefficient R_H is of the order of 10^{-9} m^3/C in (K, Ba)BiO$_3$ [22] leading to $l \sim \xi_0 \sim 30$ Å. This implies that our samples are at the border of the dirty limit, in which the electronic mean free path l is smaller than the coherence length ξ_0. In this limit, a reduction of the mean free path induces an upward shift of $H_{c2}(T) \sim 1/l\xi_0$. However, in our case, a significant shift of H_{Cp} is already observed for $B_\Phi = 1$ T, i.e., for an average distance between tracks $d \sim 400$ Å $\gg l$. Moreover, if the irradiation had induced some significant change in the mean free path, we would not have recovered the position of the pristine sample for $H\perp$ tracks. Finally, despite a large reduction of T_c we did not observe any increase of H_{Cp} in the electron irradiated sample. These arguments rule out the possibility that a decrease of the mean free path may explain the observed behavior.

Another scenario could have been that the increase of H_{Cp} is related to some local superconductivity along the tracks as observed in Al-hole networks [23]. However, in our case, the size of the hole $c_0 \sim \xi_0$ and this would lead to only a small increase of H_{c2} [24] close to T_c. Moreover, as the volume of the superconducting surface in the $B_\Phi =$ 1 T sample would be quite small, we should then have observed some additional small superconducting anomaly at $H^*_{c3} > H_{c2}$ instead of a gradual shift of the whole anomaly.

Our data strongly suggest that the specific heat anomaly is directly related to the melting of the Bose glass. However, the shape of this anomaly looks much more like the discontinuity observed at the transition towards the normal state in conventional superconductors than the one observed at the transition between different vortex states in cuprates [25]. Within this interpretation, the absence of any secondary specific heat anomaly above the IRL would imply that the vortex liquid gradually transforms into the normal state without a phase transition. The nature of the state above H_{C_p} and the role of thermal and/or quantum fluctuations in this transition still has to be clarified.

To conclude, (K, Ba)BiO_3 is a very interesting system in which $H_{Cp}(T)$ shows an anomalous positive curvature and is significantly smaller than the field H_Δ deduced from spectroscopic measurements [4]. We show here that H_{Cp} is also shifted towards higher temperature in the presence of columnar defects aligned with the external field. The observation that the specific heat anomaly follows the phenomenology of the Bose-glass melting casts serious doubts on our current understanding of the vortex liquid phase in (K, Ba)BiO_3—and probably also in other high T_c cuprates. Indeed, the strong similarities in the transport data obtained in KBBO and high T_c oxides [18,21] is suggesting that similar effects might be observed in those systems but, to the best of our knowledge, no specific heat data have been published on irradiated cuprates yet.

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